

Hysteretic and chaotic dynamics of viscous drops in creeping flows with rotation

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It has been shown in our previous publication (Bławdziewicz *et al.* 2003) that high-viscosity drops in two dimensional linear creeping flows with a nonzero vorticity component may have two stable stationary states. One state corresponds to a nearly spherical, compact drop stabilized primarily by rotation, and the other to an elongated drop stabilized primarily by capillary forces. Here we explore consequences of the drop bistability for the dynamics of highly viscous drops. Using both boundary-integral simulations and small-deformation theory we show that a quasi-static change of the flow vorticity gives rise to a hysteretic response of the drop shape, with rapid changes between the compact and elongated solutions at critical values of the vorticity. In flows with sinusoidal temporal variation of the vorticity we find chaotic drop dynamics in response to the periodic forcing. A cascade of period-doubling bifurcations is found to be directly responsible for the transition to chaos. In random flows we obtain a bimodal drop-length distribution. Some analogies with the dynamics of macromolecules and vesicles are pointed out.

1. Introduction

Investigations of dynamical properties of fluid–fluid dispersions (e.g., emulsions (Borwankar & Case 1997; Mason 1999) and polymer blends (Tucker III & Moldenaers 2002; Windhab *et al.* 2005)) require detailed understanding of the behavior of viscous drops in creeping flows. Such understanding is also crucial in development of new drop-based microfluidic systems (Whitesides & Stroock 2001; Tan *et al.* 2004; Song *et al.* 2006; Grigoriev *et al.* 2006). Therefore, drop dynamics at small Reynolds numbers have been extensively studied experimentally (Torza, Cox & Mason 1972; Bentley & Leal 1986; Bigio, Marks & Calabrese 1998; Guido, Minale & Maffettone 2000; Cristini, Guido, Alfani, Bławdziewicz & Loewenberg 2003b; Guido, Grosso & Maffettone 2004), computationally (Rallison & Acrivos 1978; Kennedy *et al.* 1994; Cristini *et al.* 1998; Zinchenko *et al.* 1999; Cristini *et al.* 2001, 2003b; Renardy 2006) and theoretically (Barthès-Biesel & Acrivos 1973; Rallison 1980; Bławdziewicz *et al.* 2002, 2003; Vlahovska *et al.* 2005).

These investigations revealed complex nonlinear drop dynamics resulting from the coupling between the drop shape and fluid flow. Examples of nonlinear phenomena that occur under creeping-flow conditions include formation of self-similar neck regions during a drop breakup process (Bławdziewicz *et al.* 1997; Lister & Stone 1998), universal slow evolution of drops near the critical flow strength above which there are no stationary drop

shapes (Bławdziewicz *et al.* 1998; Navot 1999; Bławdziewicz *et al.* 2002), and existence of two branches of stable stationary shapes of highly viscous drops in two-dimensional Stokes flows with nonzero vorticity (Bławdziewicz *et al.* 2003).

As revealed by the analysis presented by Bławdziewicz *et al.* (2003), there exists a flow-parameter range where a high-viscosity drop can either adopt a nearly spherical shape stabilized primarily by the rotational flow component or an elongated shape stabilized primarily by capillary forces. Abrupt changes of the drop shape from one state to the other can be used in manipulation of emulsion micro-structure and for controlling the behavior of highly viscous drops in microfluidic devices. Due to discontinuous changes of emulsion microstructure, the bistable drop behavior may also significantly affect emulsion rheology. Moreover, the mechanism of the bistability is of fundamental interest because of close analogies to the dynamics of vesicles (Misbah 2006; Mader *et al.* 2006; Vlahovska & Gracia 2007) and macromolecules (Bławdziewicz 2006) in external flows.

Due to the fundamental significance and because of potential applications (such as those mentioned above) it is important to explore the dynamics of highly viscous drops in linear flows with nonzero vorticity. However, the investigations presented by Bławdziewicz *et al.* (2003) were limited to stationary drop shapes and stationary external flows. In the present study we focus on drop behavior in time-dependent flows. We elucidate the physical mechanism that give rise to the bistable drop behavior and examine the consequence of these mechanism for drop response to time variation of the fluid vorticity.

The system dynamics is investigated via direct boundary-integral simulations (Pozrikidis 1992; Cristini *et al.* 2001; Bławdziewicz 2006) and by using a small-deformation approach Vlahovska (2003); Vlahovska, Bławdziewicz & Loewenberg (2005). In particular we show that the small-deformation equations with only several essential terms retained reproduce complex dynamical features of drop evolution that are associated with drop bistability.

To emphasize important aspects of the drop dynamics we consider three flow variation protocols. In the first protocol, the vorticity is slowly increased and then decreased. We find that such quasistatic vorticity ramping gives access to both the elongated and compact, nearly spherical stationary drop shapes. The drop exhibits a hysteretic behavior, with transitions between the compact shape (rotationally stabilized) and elongated shape (stabilized by capillary forces) occurring at different values of the vorticity when it is slowly ramped up or down.

In the second protocol, the vorticity undergoes finite-frequency periodic oscillations. As expected, at low frequencies the drop behavior is quasistatic, with a hysteresis loop analogous to the one observed for linear ramping. At high frequencies the oscillations average out, and the drop undergoes small oscillations around the stationary shape corresponding to the average flow. However, at intermediate frequencies we find a much more complex behavior. In particular we show that there exists a frequency and amplitude domain where the drop response to the periodic forcing is chaotic. Since in the creeping flow regime fluid motion is governed by the linear Stokes equations, the nonlinear chaotic behavior of the drop stems entirely from the coupling of the drop shape to the fluid velocity. An analysis of drop motion in the chaotic domain indicates that the transition to chaos is associated with the existence of two stationary states observed in steady flow.

In our third flow-variation protocol, the vorticity of the imposed flow undergoes random changes. We observe, that the resulting statistical distribution of the drop length is bimodal in a certain regime of flow parameters, with two peaks around the drop length corresponding to the short and long stationary solutions. Hence, we find that also in

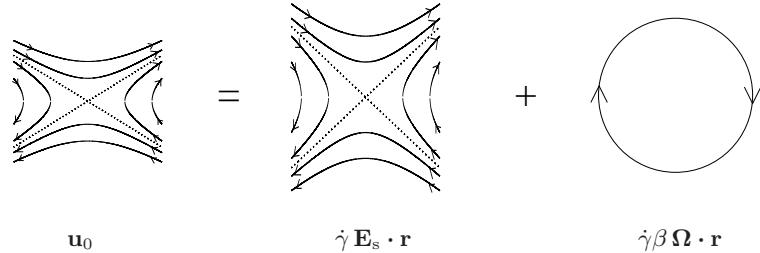


FIGURE 1. Decomposition of a linear incident flow into pure strain and rigid-body rotation.

in this problem the existence of two stationary states underpins drop behavior in a time-dependent flow.

In § 2 the system considered in our paper is defined. The quasistatic hysteretic drop behavior is analyzed in § 3, our results for chaotic drop dynamics are presented in § 4, and drop motion in linear flows with randomly varying vorticity is discussed in § 5. Our findings are summarized in § 6.

2. Viscous drops in two-dimensional linear flows

We consider a viscous drop suspended in an incompressible Newtonian fluid of a constant viscosity μ . The viscosity of the drop fluid is $\hat{\mu} = \lambda\mu$, and the interfacial tension between the two phases is σ . The drop is surfactant free, and no Marangoni stresses are present. There are also no buoyancy forces. We focus here on nonlinear effects that stem entirely from the coupling of the fluid flow to the drop shape (but not from the fluid inertia). Therefore, the creeping-flow conditions are assumed.

In the creeping-flow regime the fluid motion in the regions inside ($\mu_i = \hat{\mu}$) and outside ($\mu_i = \mu$) the drop is governed by the Stokes equations

$$\mu_i \nabla^2 \mathbf{u} = \nabla p, \quad (2.1)$$

$$\nabla \cdot \mathbf{u} = 0. \quad (2.2)$$

The fluid velocity \mathbf{u} is continuous at the drop interface Ω . Due to the absence of the Marangoni stresses the tangential viscous traction is also continuous. The jump in the normal viscous traction across Ω is equal to the capillary pressure

$$[\hat{\mathbf{n}} \cdot \boldsymbol{\tau} \cdot \hat{\mathbf{n}}] = 2\kappa\sigma, \quad (2.3)$$

where $\boldsymbol{\tau}$ is the viscous stress tensor, $\hat{\mathbf{n}}$ is the outward normal unit vector, and κ is the local curvature.

The drop is subject to two-dimensional linear incident flow

$$\mathbf{u}_0(\mathbf{r}) = \dot{\gamma}(\mathbf{E}_s + \beta\boldsymbol{\Omega}) \cdot \mathbf{r}, \quad (2.4)$$

where $\dot{\gamma}$ is the strain rate, β is the dimensionless vorticity parameter, \mathbf{r} is the position, and \mathbf{E}_s and $\boldsymbol{\Omega}$ are the symmetric and antisymmetric parts of the normalized velocity-gradient tensor. In an appropriately adopted coordinate system we have

$$\mathbf{E}_s = \frac{1}{2} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \boldsymbol{\Omega} = \frac{1}{2} \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad (2.5)$$

without a loss of generality. Accordingly, $\beta = 0$ corresponds to a purely straining flow with

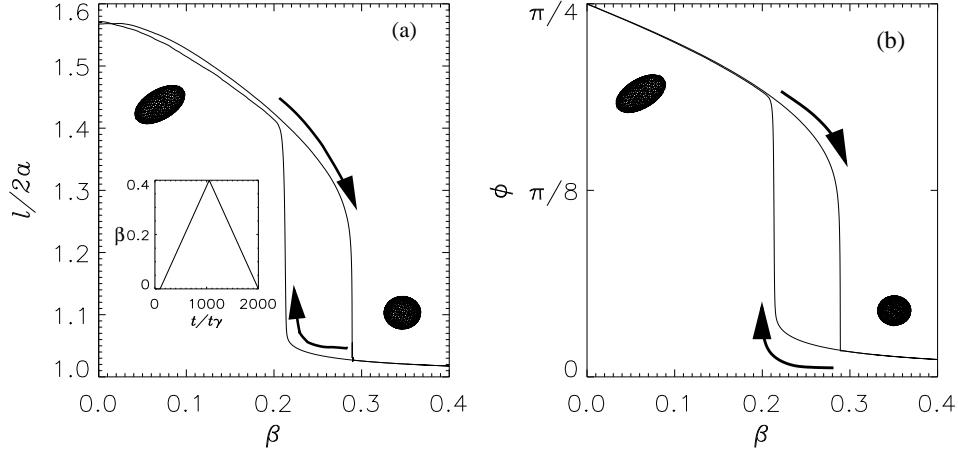


FIGURE 2. Hysteretic evolution of viscous drop in two-dimensional straining flow with slowly varying vorticity. Viscosity ratio $\lambda = 200$ and capillary number $\text{Ca} = 0.20$. (a) Normalized drop length l and (b) drop angle ϕ versus vorticity parameter β . Arrows indicate the direction of increasing time, and inset shows dependence of β on time. (Results from boundary-integral simulations.)

the extensional axis $x = y$ and the compressional axis $x = -y$, and $\beta = 1$ corresponds to shear flow in the x direction with the velocity gradient in the y direction. The tensor Ω in equation (2.4) describes the rigid-body rotation in the anti-clockwise direction with the angular velocity

$$\omega = \frac{1}{2}\beta\dot{\gamma}. \quad (2.6)$$

The decomposition of the flow field (2.4) into the straining and rotational components (2.5) is sketched in figure 1.

The dynamics of our system is characterized by three dimensionless parameters: There is the viscosity ratio λ that characterizes dissipative forces in the drop- and continuous-phase fluids. The capillary number

$$\text{Ca} = \frac{a\mu\dot{\gamma}}{\sigma} \quad (2.7)$$

(where a is the radius of an undeformed drop) characterizes the ratio between the deforming viscous forces produced by the imposed flow (2.4) and the capillary forces that resist drop deformation and drive the drop towards the equilibrium spherical shape. Finally the vorticity parameter β characterizes the magnitude of the rotational component of the external flow relative to the extensional component.

In this paper we focus on the parameter regime where the drop deformation may be significant, which requires that $\text{Ca} = O(1)$. (However, the flow is not strong enough to cause drop breakup.) We also assume that the drop-phase fluid is much more viscous than the continuous-phase fluid,

$$\lambda \gg 1. \quad (2.8)$$

Drop deformation process is hindered at large drop-phase viscosities, while drop rotation is only weakly affected by the viscous stresses inside the drops. Therefore the relative effect of the drop rotation is amplified in the regime (2.8), and the rotational component of the external flow produces nontrivial qualitative effects.

3. Hysteretic drop behavior

3.1. Capillary and rotational stabilizing mechanisms

To illustrate the effect of the rotational component of the flow (2.4) on the dynamics of a highly viscous drop we consider a system where the parameter β is first slowly increased and then slowly decreased. We adopt here a linear ramping protocol where the vorticity is slowly ramped up from $\beta = 0$ to $\beta = 0.4$ and then ramped down back to zero. Before the ramping occurs the flow is maintained at $\beta = 0$ (for 5 % of the total ramping time) to allow the drop to relax to the stationary shape in purely straining flow. This vorticity variation protocol is represented in the inset of figure 2(a).

The evolution of the drop shape in this time-dependent flow is depicted in figure 2. Figure 2(a) represents the drop length l , and figure 2(b) shows the drop angle ϕ (measured anticlockwise from the axis x); both quantities are plotted versus the vorticity parameter β . In our example, the drop viscosity is $\lambda = 200$. The capillary number $\text{Ca} = 0.2$ is below the critical value for drop breakup ($\text{Ca} = 0.22$ in 2d straining flow) but it is sufficiently large to allow for a significant flow-induced drop deformation. The total ramping time is $T = 2000t_\gamma$, so that the drop response to the flow variation is nearly quasistatic. The calculations were performed using the adaptive boundary-integral procedure developed by Cristini *et al.* (2001, 1998).

The results shown in figure 2 indicate that the drop response to the vorticity variation is hysteretic. At $\beta = 0$ drop is elongated, and it is aligned with the extensional axis of the straining component of the flow ($\phi = \pi/4$). With increasing β , the drop orientation slowly changes towards the symmetry axis x ($\phi = 0$), and the drop length slowly decreases. At a critical value of the vorticity parameter, $\beta_2 \approx 0.29$, a discontinuous change occurs: the drop length and the angle suddenly decrease. Afterwards the drop is almost spherical and nearly aligned with the axis x . When the direction of the vorticity change is reversed, the drop initially retraces its trajectory. However, the drop does not jump back to the elongated shape until the vorticity reaches the lower critical value $\beta_1 \approx 0.22 < \beta_2$.

The bistable drop behavior and the associated hysteretic shape evolution stem from the existence of two mechanisms that can stabilize a viscous drop in linear flows with rotation (2.4). Namely, the drop can be stabilized by the capillary stresses (which drive the drop towards the equilibrium spherical shape) or by the vorticity flow component (which rotates the drop out of the extensional axis of the straining component of the flow).

In a purely straining flow the drop assumes the interfacial-tension-stabilized elongated shape (Taylor 1934). The form of the drop results from the balance between drop deformation by the flow and drop relaxation due to the capillary forces. The deformation and relaxation occur on the respective time scales

$$t_\gamma = \lambda \dot{\gamma}^{-1}, \quad (3.1)$$

$$t_\sigma = \lambda \mu a \sigma^{-1}, \quad (3.2)$$

both of which are proportional the viscosity ratio $\lambda \gg 1$. The drop deformation $D = (l - 2a)/a$ is determined by the time scale ratio

$$D \sim t_\sigma/t_\gamma = \text{Ca}, \quad (3.3)$$

and therefore it is independent of the viscosity ratio in the limit $\lambda \rightarrow \infty$. In purely straining flows, the drop is oriented along the extensional axis $x = y$.

For small values of β , the vorticity flow component produces an $O(\beta)$ perturbation of the drop orientation. The corresponding decrease of the drop length is $O(\beta^2)$. However, a further drop rotation is arrested because the straining component of the flow produces

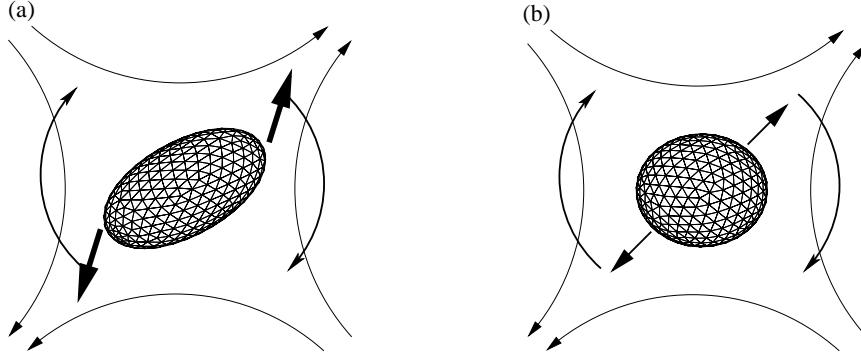


FIGURE 3. Schematic representation of the physical mechanism leading to bistable drop behavior in two-dimensional linear flows with non-zero vorticity. (a) Elongated drop is stabilized by capillary forces and destabilized by flow rotation; (b) compact drop is stabilized by flow rotation and destabilized by extensional flow component.

hydrodynamic stresses that pull the elongated drop back towards the straining axis (as illustrated in figure 3a).

Since the $O(\lambda^{-1})$ internal circulation inside an elongated high-viscosity drop is weak, the drop in its stationary state behaves analogously to a rigid body whose equilibrium orientation results from the balance of the torques produced by the straining and rotational components of the external flow. The transition to the compact drop shape occurs when the vorticity flow component becomes too strong to be balanced. Under such conditions, a rigid body would undergo a transition to a periodic motion with continuous rotation in the clockwise direction. Similarly, a drop also starts to continuously rotate when β achieves the upper critical value β_2 . However, during the rotation the drop length decreases because the drop becomes misaligned with the extensional axis of the flow. As a result, the drop relaxes to a nearly spherical shape.

In this new, compact shape the fluid inside the drop circulates with the angular velocity ω_d that is nearly equal to the angular velocity of the external flow (2.6). Within each period of rotation the drop undergoes a small deformation produced by the straining component of the external flow (as schematically illustrated in figure 3b). However, the deformation does not accumulate because it is constantly convected away by the rotational component of the flow.

Since the rotation occurs on the time scale

$$t_{\text{rot}} = (\beta\dot{\gamma})^{-1}, \quad (3.4)$$

and the drop deforms on the much longer timescale (3.1), we find that the drop deformation

$$D \sim t_{\text{rot}}/t_{\gamma} = (\beta\lambda)^{-1} \quad (3.5)$$

is small for $\lambda \gg 1$, consistent with the results shown in figure 2(a).

Relation (3.5) indicates that the deformation of the rotationally stabilized drop increases with the decreasing parameter β . When β falls below the lower critical value β_1 , the hydrodynamic torque associated with the straining component of the flow acting on a slightly elongated drop becomes strong enough to reorient the drop along the straining axis and arrest further drop rotation. Deformation thus starts to accumulate, the drop is stretched, and a transition to the interfacial-tension stabilized elongated state takes place.

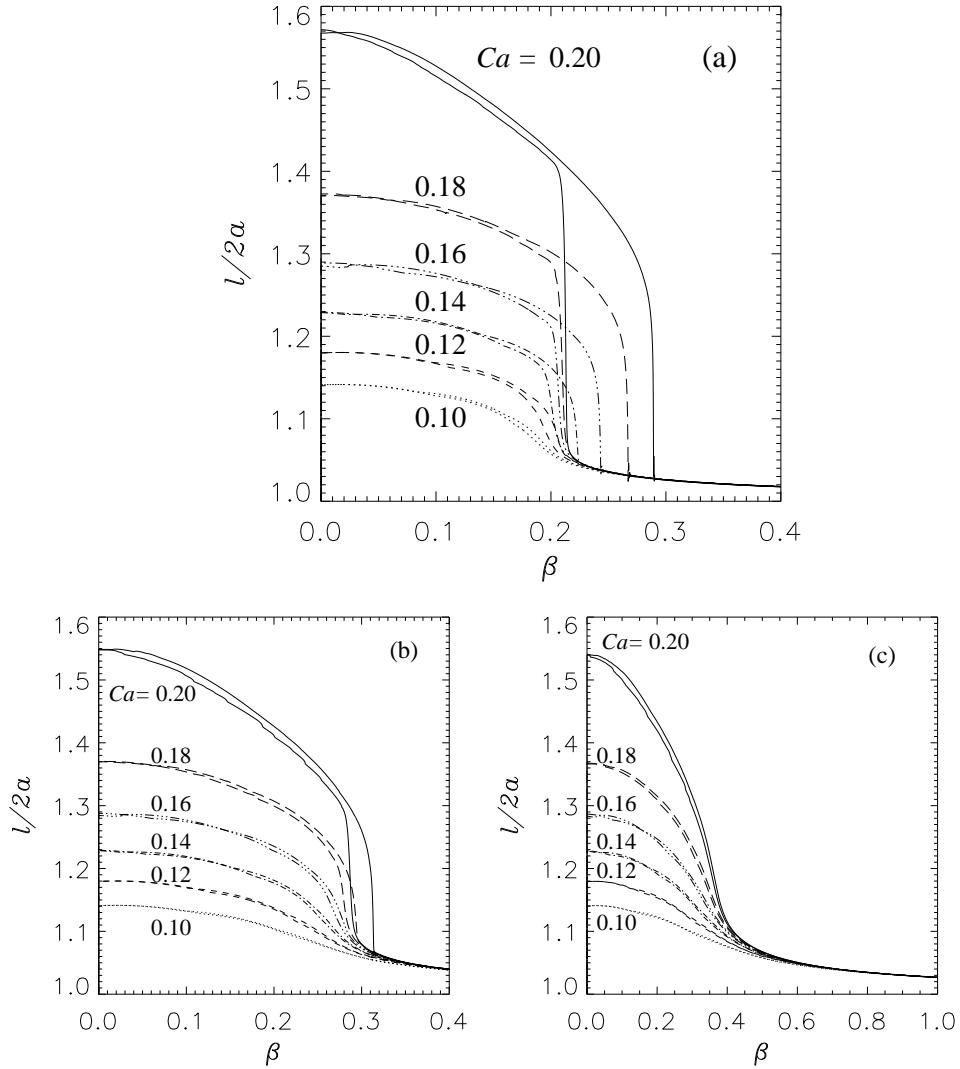


FIGURE 4. Quasistatic variation of drop length l with vorticity parameter β for (a) $\lambda = 200$, (b) 100, (c) 50, and different values of capillary number (as labeled). (Results from boundary-integral simulations.)

As shown in figure 2, a drop in the compact, rotationally stabilized stationary state is nearly aligned with the symmetry axis of the applied flow x . This behavior stems from the flow-reflection symmetry of Stokes equations and the fact that the drop is stabilized by rotation rather than the capillary forces. In the absence of the capillary forces (or in the limit of infinitely strong flow) the symmetry of Stokes equations implies that the stationary drop shape is invariant with respect to flow reflection. Hence, the shape is also invariant with respect to the corresponding transformation $(x, y, z) \rightarrow (-x, y, z)$ of the spatial coordinates (and this symmetry corresponds to drop alignment in the x direction). A perturbation due to the presence of the capillary stresses produces only a small asymmetry because the effect of capillary forces is insignificant for a nearly spherical drop.

3.2. Parameter dependence of drop response

The quasistatic response of the drop length to the variation of the vorticity is illustrated in figure 4 for different values of the capillary number Ca and viscosity ratio λ . The results indicate that the size of the hysteresis loop is the largest for large values of λ and Ca . When the capillary number is decreased, the upper critical vorticity parameter β_2 (corresponding to the transition from the elongated to the compact drop) decreases but the lower critical parameter β_1 remains nearly unaffected. In contrast, the viscosity ratio λ affects primarily the position of the lower critical parameter β_1 (corresponding to the transition from the compact to the elongated drop).

This behavior is consistent with the scaling relations (3.3) and (3.5) and the mechanism of drop bistability explained in § 3.1. According to these relations the deformation of an elongated drop scales with Ca and is independent of λ , while the deformation of a compact drop is independent of Ca and scales with λ^{-1} . We recall that the critical states corresponding to the transitions between the elongated and compact drop shapes correspond to the points where the maximal torque τ_γ exerted by the extensional component of the flow on the drop just before the transition marginally balances the torque τ_{rot} exerted by the vorticity flow component. Since $\tau_\gamma \sim D$ whereas τ_{rot} is approximately independent of D , we conclude that β_1 varies with λ^{-1} and β_2 with the Ca . This conclusion is consistent with our simulation results.

The plots shown in figure 4 indicate that for a given viscosity ratio, there exists a critical value of the capillary number Ca^* below which the drop response to the changes of the flow vorticity does not exhibit a hysteretic loop. The bifurcation point occurs at the critical value of the vorticity parameter β^* that corresponds to the position of the infinitesimal hysteresis loop for Ca slightly above the critical value Ca^* . It can be shown from the results derived by Bławdziewicz *et al.* (2003) that

$$\text{Ca}^* = \frac{15}{19} \left(\frac{5}{\lambda} \right)^{1/2}, \quad \beta^* = \frac{3}{4} \left(\frac{15}{\lambda} \right)^{1/2} \quad (3.6)$$

for $\lambda \gg 1$. The plots shown in figure 4 are consistent with the above expressions. We note that for $\lambda = 50$ (see figure 4c) the hysteretic drop behavior is not observed. The reason is that in this viscosity range Ca^* exceeds the critical value of the capillary parameter for drop breakup. Hence, the elongated stationary drop shape does not exist for $\text{Ca} > \text{Ca}^*$.

3.3. Small-deformation analysis

3.3.1. Evolution equations

Crucial features of the evolution of a highly viscous drop in two-dimensional flows with nonzero vorticity are captured by small-deformation equations. In our approach (Bławdziewicz *et al.* 2003), the position r_S of the drop interface is expanded in spherical harmonics

$$r_S/a = \alpha' + \sqrt{2} \sum_{l,m} [f'_{lm} \text{Re}(Y_{lm}) + f''_{lm} \text{Im}(Y_{lm})], \quad (3.7)$$

where $l > 0$ and $l \geq m \geq 0$ denote the spherical-harmonic order, f'_{lm} and f''_{lm} are the expansion coefficients, and the parameter α' is given by the drop-volume constraint. Since for $m = 0$ all spherical harmonics are real, we set $f''_{l0} = 0$ in expansion (3.7). Moreover, flow-induced drop deformation preserves the symmetry of the incident flow (2.4). Therefore only even values of l and m need to be included in the analysis.

Evolution equation for the expansion coefficients f'_{lm} and f''_{lm} are obtained by inserting the series (3.7) into the boundary-value problem (2.1)–(2.5), performing a boundary-

perturbation analysis, and reexpanding resulting products of spherical harmonics using appropriate Clebsch–Gordan coupling coefficients. The detailed analysis and explicit expressions for the evolution equations at different truncation levels are presented elsewhere (Vlahovska 2003; Vlahovska *et al.* 2005).

For simplicity, our small-deformation calculations are performed with the expansion (3.7) truncated at the lowest spherical-harmonic order $l = 2$, which leaves us with three independent drop-shape components: f'_{22} , f''_{22} , and f'_{20} . Noting that

$$\text{Re}(Y_{22}) \sim \cos 2\phi, \quad \text{Im}(Y_{22}) \sim \sin 2\phi, \quad (3.8)$$

we find that the shape parameters f'_{22} and f''_{22} correspond to the drop deformation along the symmetry axis x and the straining axis $x = y$, respectively. The parameter f'_{20} described an axisymmetric deformation along the axis z .

The evolution equations for the shape parameters f'_{22} , f''_{22} , and f'_{20} , truncated at the second-order in the drop deformation, can be represented in the following form

$$\dot{f}'_{20} = \lambda^{-1} (d_{11} + d_{12} f'_{20}) f''_{22} - \lambda^{-1} \text{Ca}^{-1} [D_1 f'_{20} - D_2 (f'^2_{20} - f'^2_{22} - f''^2_{22})], \quad (3.9a)$$

$$\dot{f}'_{22} = -2\omega f''_{22} + \lambda^{-1} [d_{21} f'_{22} f''_{22} - \text{Ca}^{-1} (D_1 + 2D_2 f'_{20}) f'_{22}], \quad (3.9b)$$

$$\dot{f}''_{22} = 2\omega f'_{22} + \lambda^{-1} [(d_{31} + d_{32} f'_{20} + d_{33} f'^2_{20} + d_{34} f'^2_{22} + d_{35} f'^2_{22}) - \text{Ca}^{-1} (D_1 + 2D_2 f'_{20}) f''_{22}], \quad (3.9c)$$

where the dot denotes the time derivative (normalized by $\dot{\gamma}^{-1}$). The terms involving the coefficients d_{ij} correspond to drop deformation by the external flow, and the terms involving D_k describe the capillary relaxation. All these terms are $O(\lambda^{-1})$ in the large-viscosity-ratio regime. Explicit expressions for the coefficients d_{ij} and D_k are given in (Vlahovska 2003); here we only note that these coefficients are functions of the viscosity ratio λ and have finite limits for $\lambda \rightarrow \infty$.

The two remaining terms on the right-hand side of equations (3.9b) and (3.9c) (the terms proportional to ω) are viscosity independent. These terms represent the rigid-body rotation of the drop, with the angular velocity

$$\omega = -\frac{1}{2}\beta + \frac{1}{2}c_1 f'_{22}, \quad (3.10)$$

where $c_1 = (15/2\pi)^{1/2}$. Consistent with our qualitative physical picture described in § 3.1 (and illustrated in figure 3), the rotational velocity (3.10) involves two terms. The first term corresponds to the rotation of the drop by the vorticity component of the flow (2.4). The second term, which is proportional to the shape parameter f'_{22} that described deformation in the x direction, corresponds to the rotation of a deformed drop by the straining component of the external flow towards the straining axis $x = y$.

3.3.2. Reduced description

It has been shown by Bławdziewicz *et al.* (2003) that for $\lambda \gg 1$ the drop behavior near the bifurcation point (3.6) can be described by simplified asymptotic equations

$$\dot{f}'_{22} = -2\omega f''_{22} - \lambda^{-1} \text{Ca}^{-1} \bar{D}_1 f'_{22}, \quad (3.11a)$$

$$\dot{f}''_{22} = -2\omega f'_{22} - \lambda^{-1} \text{Ca}^{-1} \bar{D}_1 f''_{22} + \lambda^{-1} \bar{d}_{31}. \quad (3.11b)$$

where $\bar{D}_1 = 20/19$ and $\bar{d}_{31} = (5\pi/6)^{1/2}$ are the high-viscosity limits of D_1 and d_{31} . The asymptotic result (3.11) is obtained from (3.9) on assumption that near the bifurcation point there is a balance between drop deformation and rotation (which corresponds to $\lambda^{-1} \sim \omega f'_{22}$) and the balance between capillary relaxation and rotation (which yields $(\lambda \text{Ca})^{-1} f'_{22} \sim f''_{22}$ and $(\lambda \text{Ca})^{-1} f''_{22} \sim f'_{22}$). Moreover, the two contributions to the angular

velocity (3.10) are of the same order but do not cancel (i.e., $\omega \sim \beta \sim f'_{22}$). Equations (3.9) are rescaled accordingly, and only the leading order terms are retained.

Equations (3.11) have all necessary ingredients that are needed to describe the hysteretic drop behavior. We have terms representing drop rotation by the straining and vorticity components of the external flow (i.e., the terms proportional to ω), drop relaxation due to the presence of the capillary forces (the terms proportional to Ca^{-1}), and stretching of the drop along the straining axis of the straining component of the external flow (the last term in equation (3.11b)). Neglecting any of these terms would qualitatively alter the solution structure, which no longer would manifest the key features of the drop evolution in the parameter range considered herein.

3.3.3. Asymptotic solution

In the regime $\lambda^{-1} \ll 1$ and $\beta \ll 1$ the stationary solutions of equations (3.11) can be obtained by a singular-perturbation analysis. To the leading order in the small parameters, we find that the elongated drop is described by the relations

$$\omega \simeq 0, \quad (3.12a)$$

$$\lambda^{-1} \text{Ca}^{-1} \bar{D}_1 f''_{22} \simeq \lambda^{-1} \bar{d}_{31}. \quad (3.12b)$$

The first of the above relations correspond to the fact that an elongated drop does not rotate (there is only a weak fluid circulation inside it, as predicted by our qualitative analysis). The second relation describes the balance between drop deformation by the external flow and relaxation due to the capillary forces. Recalling the definition (3.10) of the angular velocity ω we find that relations (3.12) yield

$$f'_{22} \simeq c_1 \beta, \quad (3.13a)$$

$$f''_{22} \simeq \bar{D}_1^{-1} \bar{d}_{31} \text{Ca}. \quad (3.13b)$$

By inserting the above relations back into (3.11) one can verify that they constitute a consistent leading-order asymptotic stationary solution.

According to equation (3.13b) drop elongation along the straining axis $x = y$ (i.e., $\phi = \pi/4$) scales with the capillary number, which is consistent with the scaling result (3.3). The drop angle

$$\phi = \frac{1}{2} \arctan(f''_{22} / f'_{22}) \quad (3.14)$$

only slightly deviates from $\phi = \pi/4$ because $f'_{22} \ll f''_{22}$ (assuming that $\text{Ca} \gg \beta$ and $\beta \ll 1$).

The leading-order stationary solution corresponding to the compact drop is obtained from the following relations

$$-2\omega f''_{22} \simeq 0, \quad (3.15a)$$

$$2\omega f'_{22} \simeq -\lambda^{-1} \bar{d}_{31}, \quad (3.15b)$$

which are obtained by dropping from the evolution equations (3.11) the $O(\lambda^{-1})$ capillary-relaxation terms. Taking into account the definition (3.10) of ω we thus obtain

$$f''_{22} \simeq 0 \quad (3.16a)$$

and

$$f'_{22} \simeq \frac{\beta - \sqrt{\beta^2 - 4c_1 \bar{d}_{31} \lambda^{-1}}}{2c_1} \quad (3.16b)$$

(the solution with the plus sign in front of the square root is unstable).

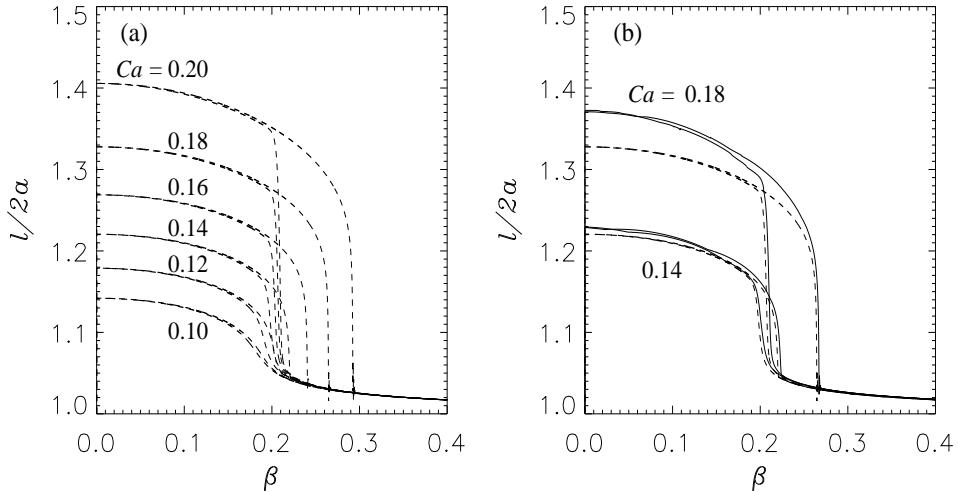


FIGURE 5. Quasistatic variation of drop length l with vorticity parameter β for $\lambda = 200$ and different values of capillary number (as labeled). (a) Solution of small-deformation equations (3.9); (b) comparison of small-deformation results (dashed lines) with boundary integral simulations (solid lines).

Since the shape parameter f''_{22} vanishes according to equation (3.16a), the drop is oriented in the x direction. For $\lambda^{-1} \ll \beta$ we find

$$f'_{22} = \bar{d}_{31} \beta \lambda^{-1}, \quad (3.17)$$

which is consistent with our scaling estimate (3.5). For $\beta < \beta_1$, where

$$\beta_1 = 2(c_1 \bar{d}_{31})^{1/2} \lambda^{-1/2}, \quad (3.18)$$

the solution (3.16b) does not exist; the drop thus undergoes a transition to the long solution. The critical vorticity parameter decreases with increasing λ , in agreement with our numerical results presented in figure 4.

The solution (3.12)–(3.18) of the simplified small-deformation equations (3.11) is perturbative. We note, however, the exact stationary solution can also be found (Bławzdziewicz *et al.* 2003). Our analytical solution quantitatively agrees with the results of numerical simulations, provided that the drop deformation is not too large.

3.3.4. Numerical results

Predictions of the small-deformation equations (3.9) for drop behavior in a two-dimensional linear flow with slowly varying vorticity are depicted in figure 5 for a system with the viscosity ratio $\lambda = 200$. Figure 5(a) shows the dependence of the drop length

$$l/a = 1 + \sqrt{\frac{15}{8\pi}} (f'^2_{22} + f''^2_{22})^{1/2} \quad (3.19)$$

on the vorticity parameter β for the same set of capillary numbers as those represented in figure 4. Figure 5(b) compares the small deformation results directly with the results of the boundary-integral simulations. The small-deformation calculations were performed using the second-order equations (3.9) because for an elongated drop they are more accurate than the simplified equations (3.12).

The results shown in figures 4 and 5 indicate that for small and moderate capillary numbers the small-deformation theory yields accurate quantitative predictions. At high

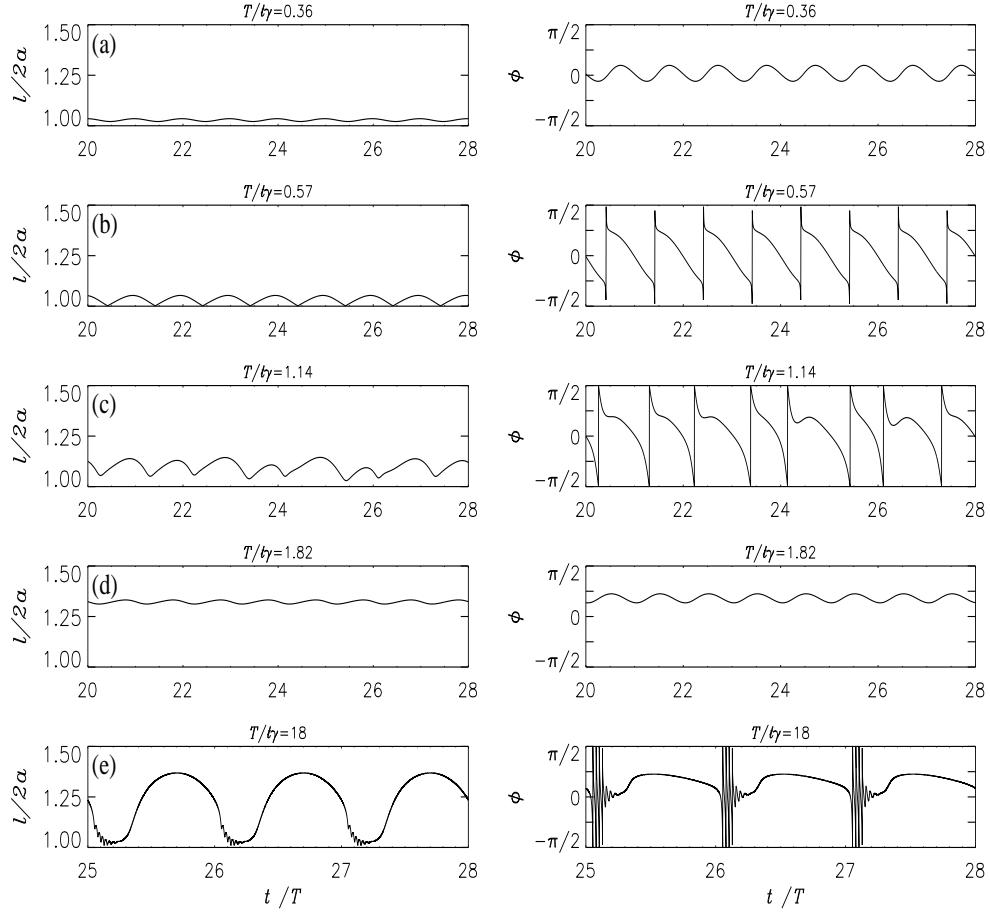


FIGURE 6. Evolution of drop length (left) and angle (right) in two dimensional linear flow with harmonic variation of vorticity (4.1), for different values of period T normalized by drop-deformation time (as labeled). Mean vorticity $\bar{\beta} = 0.25$, vorticity amplitude $\delta\beta = 0.13$, viscosity ratio $\lambda = 275$ and capillary number $\text{Ca} = 0.2$. Panels (c) depict chaotic dynamics. (Results from small-deformation theory.)

values of Ca drop behavior is also captured quantitatively, except for the upper portion of the hysteresis loop (i.e. when the drop is in the elongated state). For all values of the capillary number the lower and upper critical vorticity parameters β_1 and β_2 are obtained within the numerical error of the boundary-integral simulations. Our additional calculations (not shown) indicate that a similar accuracy is obtained for $\lambda = 50$ and $\lambda = 100$.

4. Chaotic drop dynamics in a sinusoidal straining flow

Dynamical systems with multiple equilibrium states often exhibit novel dynamics when driven by simple forcing (Guckenheimer & Holmes 1983a). Thus, despite the laminar nature of the Stokes flow, we expect to find interesting nonlinear dynamics of a viscous drop in a time-varying linear flow with rotation. To explore this dynamics we will now

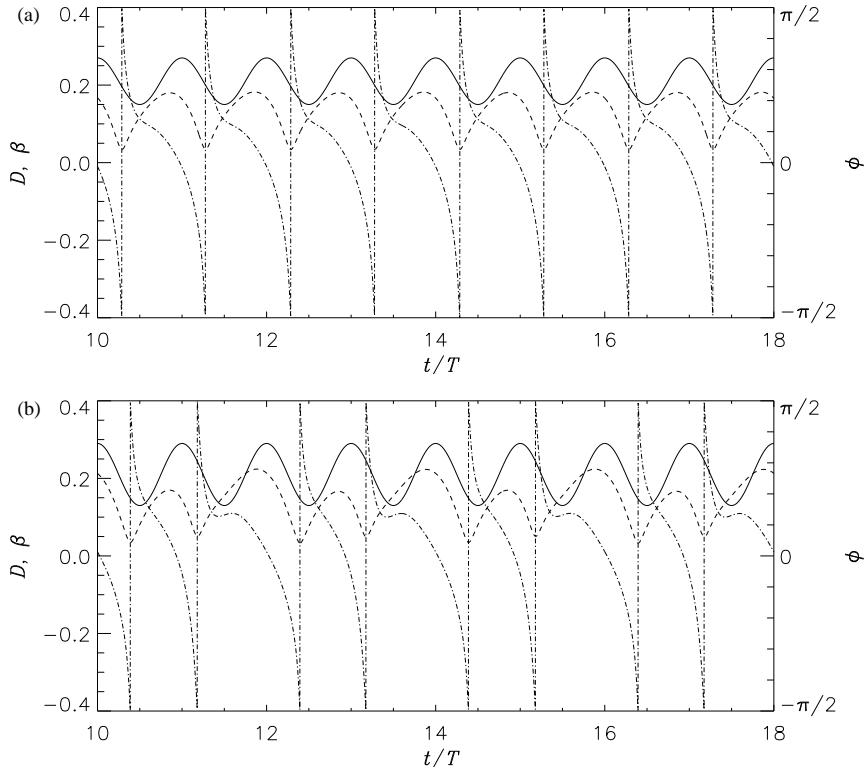


FIGURE 7. Period doubling in the dynamics of viscous drop in two-dimensional linear flow with harmonic variation of vorticity. Vorticity parameter β (solid lines), drop deformation D (dashed), and drop angle ϕ (dash-dotted) are shown versus time t normalized by the oscillation period T . Viscosity ratio $\lambda = 275$, capillary number $\text{Ca} = 0.2$, period $T/t_\gamma = 1.14$, mean vorticity $\bar{\beta} = 0.21$, and vorticity oscillation amplitude (a) $\delta\beta = 0.6$ and (b) 0.8 . (Results from small-deformation theory.)

investigate the drop response to harmonic variation of the vorticity

$$\beta(t) = \bar{\beta} + \delta\beta \cos(2\pi t/T), \quad (4.1)$$

where $\bar{\beta}$ is the average vorticity value, $\delta\beta$ is the oscillation amplitude, and T is the oscillation period.

We have performed a series of small deformation calculations (§ 4.1) and boundary-integral simulations (§ 4.2) for different values of the flow parameters $\bar{\beta}$, $\delta\beta$, and T . If the oscillation period T is much shorter than the drop deformation and oscillation times (3.1) and (3.2), we find that the drop undergoes small oscillations about a stationary shape corresponding to the mean value of β (which is an expected behavior). In the opposite regime $T \gg t_\gamma, t_\sigma$, the quasistatic drop behavior described in § 3 is recovered. In what follows we focus on the most interesting parameter domain $T \sim t_\gamma, t_\sigma$ and $\delta\beta \sim \beta_1 - \beta_2$, in which an interaction of different timescales as well as an interplay between the short and elongated drop shapes is anticipated.

4.1. Small-deformation results

Figure 6 illustrates the dependence of the drop evolution in linear flow with the oscillatory vorticity (4.1) on the oscillation period T . The viscosity ratio in this example is $\lambda = 275$,

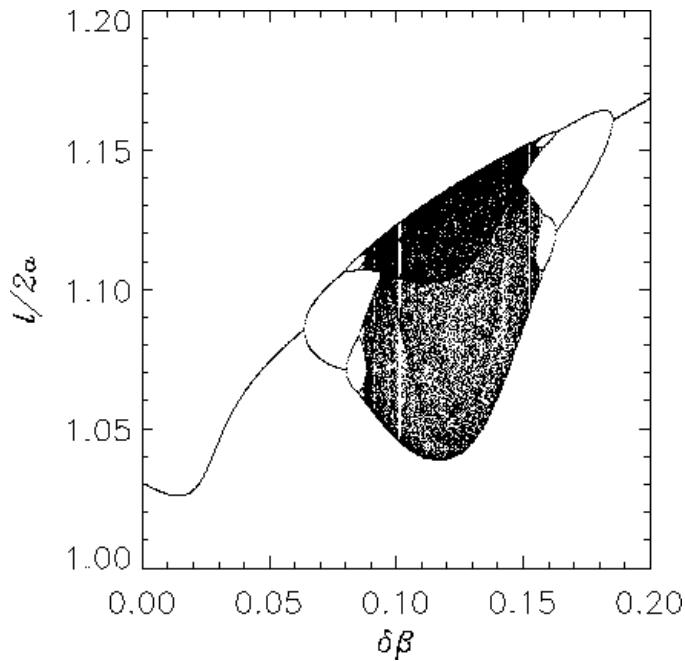


FIGURE 8. Bifurcation diagram showing period doubling cascades and transition to chaos for viscous drop in linear flow with harmonic vorticity variation with mean $\bar{\beta} = 0.21$ and period $T/t_\gamma = 1.14$. Viscosity ratio and capillary number are the same as in figures 6 and 7. (Results from small-deformation theory.)

and the capillary number is $Ca = 0.2$. The mean value of the vorticity $\bar{\beta} = 0.21$ is close to the lower critical value $\beta_1 = 0.18$, and the oscillation amplitude is $\delta\beta = 0.13$.

Figure 6(a) represent our results for the shortest oscillation period of the flow vorticity $T/t_\gamma = 0.36$. The drop oscillates about the compact stationary shape in this case. Both the drop lengths and the drop angle vary periodically, with the period T_d equal to the period T of the external forcing. The drop length decreases when the drop is in the compressional quadrant $-\pi/2 < \phi < 0$ and increases for $0 < \phi < \pi/2$. Upon an increase of the period of the external forcing the amplitude of the angular drop oscillations increases. When the oscillation amplitude reaches $\pi/2$ the drop starts to tumble, as illustrated in figure 6(b).

A further increase of the period of the external forcing results in a qualitative change of drop response. We find that the drop still undergoes a tumbling motion; however, the evolution is not periodic but it becomes *chaotic*, as shown in figure 6(c). The chaotic motion continues up to $T/t_\gamma = 1.14$, and then the drop reverts to periodic motion. For T/t_γ (figure 6d) the drop oscillates about the elongated stationary shape, and in the regime $T/t_\gamma \gg 1$ (figure 6e) the system approaches the quasistatic behavior discussed in § 3.†

The transition to the chaotic drop motion occurs through a cascade of period doubling events, as illustrated in figures 7 and 8. Figure 7 depicts the drop evolution at three values of the amplitude of vorticity oscillations $\delta\beta$ (the remaining system parameters are

† The only significant deviation from the quasistatic evolution for $T/t_\gamma \gg 1$ occurs right after the drop jumps from the long to the compact shape when β increases above the upper critical value β_2 . Namely, before the drop settles down to the compact stationary shape it undergoes a tumbling motion with the amplitude decaying on the timescale (3.2).

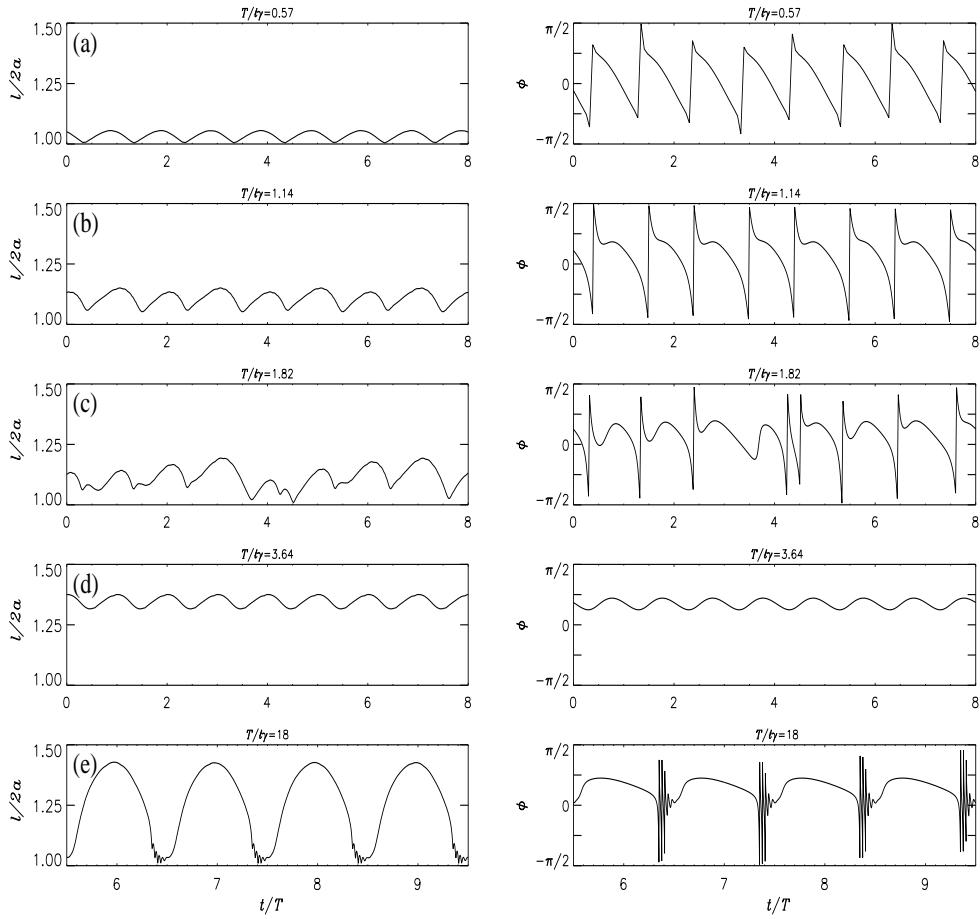


FIGURE 9. Same as figure 6, except that the results are from boundary-integral simulations and for slightly different values of oscillation period. Same $\bar{\beta}$ and $\delta\beta$ as in figure 6.

the same as those that yield the chaotic motion depicted in figure 6(c). The results shown in figure 7(a) indicate that for a sufficiently small oscillation amplitude ($\delta\beta = 0.06$ in our example) the drop evolves with the period $T_d = T$ (i.e., the period equal to that of the external forcing). At a larger amplitude $\delta\beta = 0.08$ the drop oscillation period is $T_d = 2T$ and for $\delta\beta = 0.083$ we find $T_d = 4T$. The period-doubling scenario of the transition to chaos in our system is further supported by the bifurcation diagram shown in figure 8, where the drop length l at times $t = nT$ ($n = 1, 2, \dots$) is plotted versus the flow-oscillation amplitude $\delta\beta$.

An analysis of the results shown in figure 7 indicates that the period doubling occurs as a result of a resonance between the drop tumbling motion and the vorticity oscillations. Namely, if the drop is relatively long and approximately aligned with the straining axis of the external flow when the vorticity parameter $\beta(t)$ reaches a minimum, the drop rotation may be significantly slowed down or even arrested (as in the long-drop stationary state discussed in §3). Such a temporary arrest of drop rotation corresponds to the shoulders in the plots of the angular evolution depicted in figure 7. On the other hand, if the drop angle exceeds $\phi = \pi/2$ when the vorticity goes through a minimum, the arrest of drop rotation does not occur. As seen in figure 7(b) this interplay of drop tumbling with

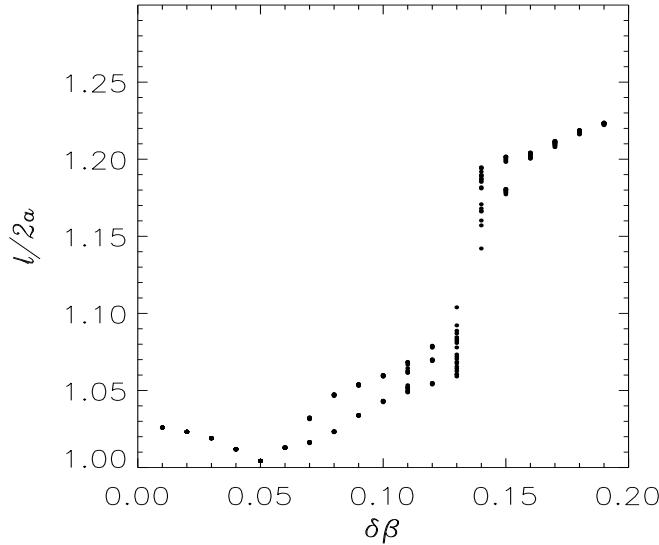


FIGURE 10. Bifurcation diagram for viscous drop in linear flow with harmonic vorticity variation with mean $\bar{\beta} = 0.22$, $T/t_\gamma = 1.82$, $\lambda = 275$, and $\text{Ca} = 0.2$. (Results from boundary-integral simulations.)

oscillations of the external forcing produces the period-doubling bifurcation that leads to alternating accelerated and retarded drop-rotation cycles.

In our numerical calculations depicted in figures 6-8 we have used the full set of the small-deformation equations (3.9) but we find that the simplified asymptotic equations (3.11) yield similar results. In particular these equations correctly reproduce the cascade of the period doubling bifurcations and the chaotic-evolution domain. However, a further simplification of the evolution equations is not possible: if any of the terms in equations (3.11) is removed the solutions qualitatively change and the chaotic domain disappears. We also find that chaotic drop dynamics occurs only for highly viscous drops with $\lambda > 200$.

4.2. Boundary-integral results

Figure 9 shows examples of our simulation results for the periodic and chaotic drop evolution. The flow parameters are similar to those used in our small-deformation calculations described in § 4.1. As with the small-deformation calculations, for short periods of the external forcing T the drop oscillates around the compact stationary shape, for moderate periods the system undergoes a transition to chaotic evolution, and for long periods the drop motion approaches the quasistatic behavior. Consistent with the small-deformation results, the chaotic drop dynamics revealed by the boundary-integral simulations is due to a cascade of period doubling bifurcations. A bifurcation diagram illustrating this behavior is presented in figure 10.

The domain of chaotic motion found in the direct boundary-integral simulations somewhat differs from the corresponding domain obtained from the small-deformation theory. Also, the magnitude of the chaotic fluctuations in the drop length is larger in the boundary-integral runs. We have tested the convergence of the boundary-integral simulations, and we believe that the differences between the drop behavior obtained by the two different methods stem from the approximations involved in the small-deformation theory. We note, however, that the evolution in the chaotic and period-doubling regimes

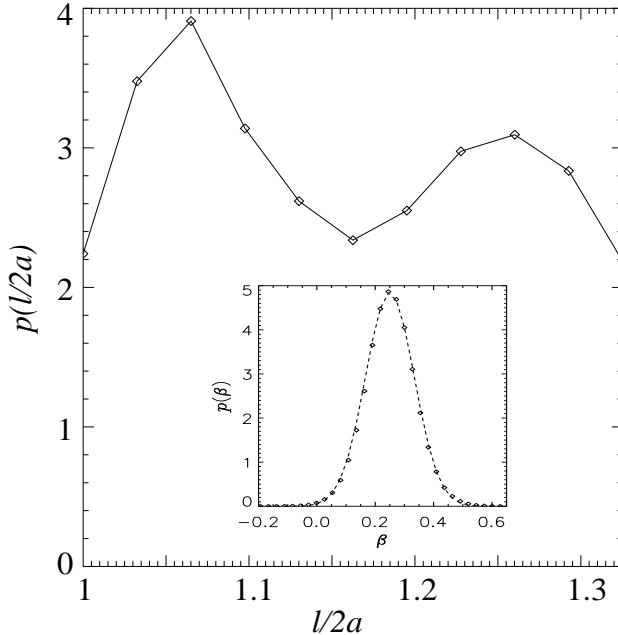


FIGURE 11. Length probability distribution for viscous drop with $\lambda = 200$ and $\text{Ca} = 0.2$, in linear flow with stochastic vorticity. Mean value of the vorticity parameter $\langle \beta \rangle = 0.25$, variance $\delta\beta = 0.13$, and correlation time $\tau^{\text{corr}}/t_\gamma = 0.24$. Inset shows vorticity probability distribution. (Results from boundary-integral simulations.)

is very sensitive to the to small changes of the system parameters (hence, also sensitive to the approximations involved in our calculations).

5. Drop Statistics in a linear flow with Stochastic Vorticity

In some systems (e.g. emulsion flows through a packed bed of fibers (Mosler & Shaqfeh 1997) or turbulent emulsion flows with drops that are much smaller than the Kolmogorov scale (Cristini *et al.* 2003a)) a viscous drop undergoes deformation in a random external creeping flow. To gain some understanding of the role that drop bistability may play in such systems we consider the drop behavior in a flow with stochastic vorticity.

We assume that the time variation of the vorticity parameter β is described by a stationary Markovian Gaussian process (i.e, the Ornstein–Uhlenbeck process) with the mean $\langle \beta \rangle$, variance $\delta\beta$, and correlation time τ^{corr} . A standard numerical scheme (Fox *et al.* 1988) for generating such a time-correlated Gaussian process is used to model the time variation of β for a given drop trajectory.

An example of drop behavior in a stochastic flow with a Gaussian variation of the vorticity is presented in figure 11. The mean value of the vorticity is $\langle \beta \rangle = 0.25$ and the variance is $\delta\beta = 0.13$. The correlation time of the vorticity distribution $\tau^{\text{corr}}/t_\sigma = 0.24$ is several times shorter than the drop relaxation time. The figure depicts the probability distribution of the drop length, obtained using the boundary-integral simulations. The capillary number is $\text{Ca} = 0.2$, and the drop viscosity is $\lambda = 200$.

The results indicate that the drop-length distribution is bimodal for the above parameter values. This behavior is expected since the vorticity undergoes random variation in the domain that includes both the lower and upper critical values β_1 , and β_2 of the

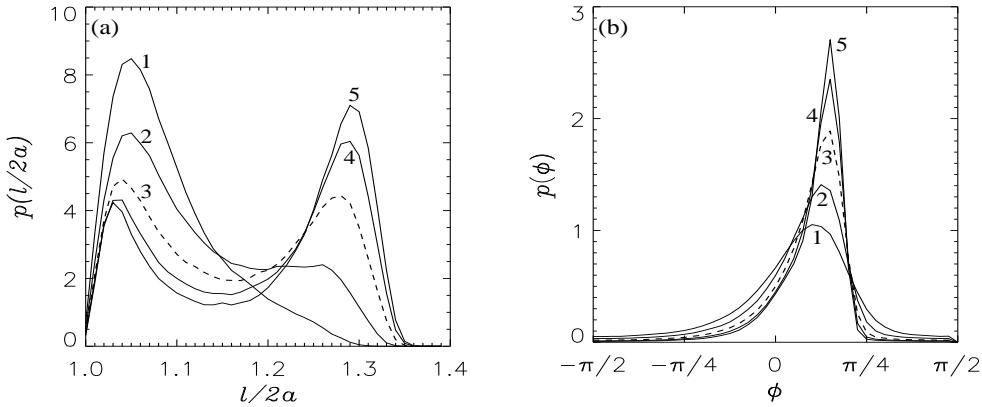


FIGURE 12. (a) Length and (b) angle probability distributions for viscous drop with $\lambda = 200$ and $\text{Ca} = 0.2$, in linear flow with stochastic vorticity. Mean and variance of the vorticity parameter are the same as in figure 11; the correlation times are $\tau^{\text{corr}}/t_\gamma = 0.025, 0.1, 0.2, 0.3, 0.4$ for lines marked 1–5, respectively. (Results from small-deformation theory.)

vorticity parameter β . Since drop response to a flow with slow variation of vorticity is hysteretic, a drop in the random flow tends to stay in the neighborhood of the compact and the elongated stationary states.

We note that the peak of the length probability distribution at $l \approx 1.25$ is shifted towards the shorter drop lengths compared to the length of a drop in the elongated stationary shape. This is because $\text{Ca} = 0.2$ is close to the critical capillary number for drop breakup. Thus, due to the slow time scale in the drop dynamics near the critical capillary number (Bławdziewicz *et al.* 2002), the drop does not have sufficient time to fully extend before the vorticity significantly changes.

Figure 12 shows the probability density distribution for the drop length l and drop angle ϕ for different values of the flow correlation time τ^{corr} . Other system parameters are the same as in figure 11. The calculations were performed using the small-deformation equations (3.9). The results indicate that at short flow correlation times the drop-length probability distribution is peaked around small values corresponding to the short-drop stationary solution, and has a moderate-height peak at $\phi \approx \pi/4$. As the flow correlation time τ^{corr} increases, the length probability distribution becomes bimodal: one of its peaks corresponds to compact and the other to elongated drops. A corresponding change occurs in the angle distribution, i.e., its peak becomes more pronounced, and shifts towards the straining axis $\phi = \pi/4$.

The shift of the typical drop length and orientation from the compact to elongated state when the flow correlation time is increased resembles the analogous shift for a system with harmonic vorticity oscillations (see figures 6b and figures 6d). This behavior is further illustrated in figure 13 which shows the average values and the variance of l and ϕ versus the correlation time τ^{corr} .

6. Conclusions

We have presented results of numerical and theoretical investigations of the dynamics of highly viscous drops in two-dimensional linear creeping flows with time-dependent vorticity. In our earlier publication (Bławdziewicz *et al.* 2003) we predicted that in stationary flows such drops exhibit bistable behavior: there is a range of system parameters

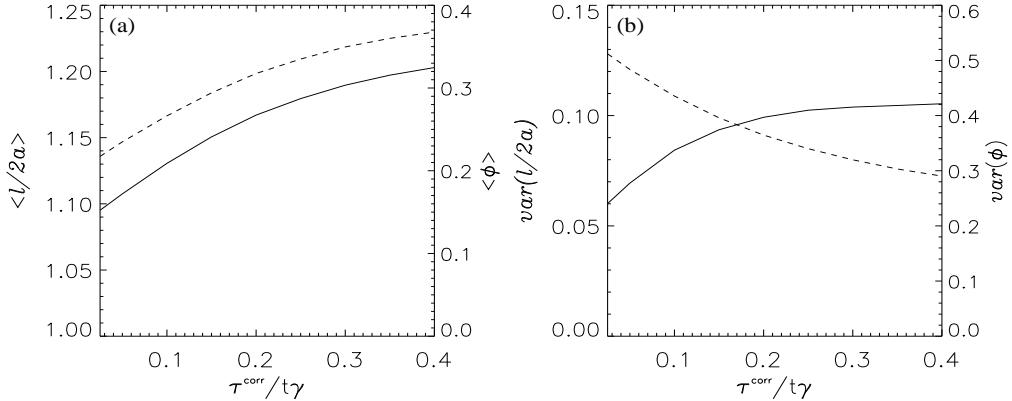


FIGURE 13. (a) Mean and (b) variance of drop length (dashed) and angle (broken line) versus correlation time τ^{corr} normalized by drop deformation time, for $\lambda = 200$, $\text{Ca} = 0.2$, $\langle \beta \rangle = 0.25$, and $\delta\beta = 0.13$.

where the drop may assume either an elongated shape approximately aligned with the straining axis of the external flow or a nearly spherical shape approximately aligned in the flow direction. Here we analyze the consequences of this behavior for the system dynamics. We also elucidate the physical mechanism that leads to drop bistability.

A direct consequence of the existence of two stationary states is hysteretic drop response to a flow with slowly varying vorticity. We have explained that the rapid transition from an elongated non-rotating drop shape to the nearly spherical compact shape occurs when the vorticity becomes strong enough to overcome the effect of the straining flow component that aligns the drop with the straining axis. This transition is thus analogous to the behavior of an elongated rigid particle which starts to tumble when the vorticity grows above a critical value (Jeffery 1922). A viscous drop also begins to tumble at a critical vorticity magnitude β_2 . However, when the drop becomes misaligned with the extensional axis it relaxes under the action of capillary forces towards a nearly spherical rotationally stabilized stationary shape.

If, in turn, the vorticity is slowly decreased, the drop returns to the elongated shape only after the vorticity magnitude reaches a lower critical value $\beta_1 < \beta_2$. This hysteretic drop response occurs at high drop viscosities because the rotational stabilizing mechanism is more efficient in the high-viscosity regime. A highly viscous drop deforms less within each drop revolution, so the compact shape remains stable even for small vorticity magnitudes.

The existence of two stationary states affects drop dynamics not only in the quasistatic regime but also at finite frequencies of the external forcing. At small amplitudes of harmonic vorticity oscillations the drop simply oscillates (with the same frequency as the external forcing) about one of the stationary states. However, if the vorticity-variation range includes both critical values β_1 and β_2 the dynamics of the system is much richer. We find that with an increasing magnitude of the vorticity oscillations the system undergoes a cascade of period-doubling bifurcations resulting in chaotic drop dynamics. The period doubling stems from the resonance between the periodicity of the external forcing and the tumbling motion of the drop when it jumps from a (partially) elongated shape towards the compact rotationally stabilized state.

Chaos in our system emerges despite linearity of Stokes equations – the system dynamics is nonlinear because of the coupling of the flow to the evolving fluid interface. A

detailed analysis of small-deformation equations describing drop dynamics reveals that in addition to chaos associated with the period-doubling mechanism there also exists in our system a different kind of chaotic evolution that results from manifold tangling (Guckenheimer & Holmes 1983b). Our analysis of different types of chaos will be presented in a separate publication.

To our knowledge, chaotic drop dynamics in Stokes-flow regime has never been observed before. We note, however, that in a recent independent study (Kas-Danouche *et al.* 2007) has reported chaotic dynamics in co-annular Stokes flow with insoluble surfactant adsorbed on the fluid interface. Chaos in their system also appears as a result of period doubling.

Understanding of drop bistability and the associated dynamical phenomena is relevant for many practical problems. For example, interpretation of rheological response of emulsions of highly viscous drops to time-varying flows requires insight into drop dynamics. Our results may also be useful in design new methods for manipulating emulsion microstructure in material processing and controlling drop behavior in microfluidic flows. Drop bistability could, e.g., be used to construct microfluidic switches, and chaotic drop dynamics may be relevant for microfluidic mixing.

The stabilizing and destabilizing mechanisms described in our paper apply not only to viscous drops but also to other deformable particles. Therefore, results of our study have a broader significance.

In particular, our analysis suggests that macromolecules with high degree of internal dissipation may undergo a transition between a nearly spherical and moderately elongated states (in addition to the standard coil-stretch transition predicted by de Gennes 1974). In fact, the dynamics of macromolecules can be modeled using equations analogous to (3.11), supplemented with terms representing random thermal forces. Such a simplified description correctly captures the most important features of power spectra of DNA molecules evolving in linear flows with nonzero rotational component (Bławdziewicz 2006).

There are also close analogies between drop and vesicle motion. The main difference between these two systems is that vesicles satisfy a constant-area constraint whereas the drop area can vary. This constraint gives rise to periodic vesicle motion (such as tank treading and tumbling) even in stationary flows (Misbah 2006; Vlahovska & Gracia 2007). It would be interesting to determine if a coupling of vesicle oscillations to a harmonic variation of the external flow can lead to chaotic dynamics.

It would also be of significant interest to experimentally explore the bistable and chaotic drop dynamics (as well as related phenomena for other deformable particles). In such experiments a four-roll mill could be used to produce a linear flow with a controlled magnitude of vorticity. The experiments could also be performed using recently developed microfluidic analogues of a four-roll mill device (Hudson *et al.* 2007; Lee *et al.* 2007).

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